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13. ABSTRACT (Maximum 200 words)

This report describes results of field and laboratory studies focusing on the Ob River system. Analyses of suspended particles collected in 1994 and 1995 show that the $^{240}Pu/^{239}Pu$ atom ratio is lower than global fallout throughout the system (except in the Taz River). Low values of the ratio in the Ob River before it joins the Irtysh suggest that Tomsk-7 is an important source of reprocessing Pu to the system. As much as 39% of the Pu in this part of the river could be derived from reprocessing. $^{239,240}Pu/^{137}Cs$ activity ratios are depressed relative to global fallout in the Irtysh and Tobal Rivers suggesting that releases from Mayak are also occurring and labelling particles in the system. Mayak is also the apparent source of elevated ^{129}I to the Ob. Decreases in ^{129}I from the Tobal to the Ob at confluence points of the Tobal with the Irtysh and Irtysh with the Ob are evident, probably due to dilution. Laboratory mixing experiments of filtered Ob River water and sea water were performed to determine radionuclide removal in the estuary of the Ob. Removal varied in the order Am>>>Co=I≥Cs. Am displayed significant removal (~38%) at 5 ppt salinity, with most of the uptake onto particles in the 0.2 - 1 μm size range. Cs and I were most reactive at 0 ppt salinity and uptake decreased with increasing salinity. Irradiation of Ob River water with UV to photooxidize dissolved organic matter caused an increase in uptake of Am onto particles.

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FINAL REPORT

**Transport and fate of anthropogenic radionuclide contaminants
in the Ob River estuarine system**

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I. OBJECTIVES AND APPROACH

The Ob river system in western Siberia drains into the Kara Sea and may represent an important pathway for the delivery of anthropogenic radionuclides to the Arctic. The Ob system has received radioactive contaminants from global fallout from nuclear weapons testing, local fallout from Novaya Zemlya and perhaps most significantly, discharges from nuclear facilities sited near the Techa, Iset and Tom rivers, which drain into the Ob. In particular, releases of radioactivity from the MAJAK installation near Chelyabinsk and from the Siberian chemical plant Tomsk-7 have contributed anthropogenic radionuclides (^{137}Cs, ^{90}Sr, ^{239,240}Pu, ^{241}Am, ^{60}Co, ^{129}I and others) to the Ob and its tributaries. Our research is focusing on the extent to which these released radionuclides can be seen in the water and suspended sediments of the Ob system.

We have conducted an integrated field and laboratory study to better understand the concentrations, transport pathways, geochemical fate and biological uptake of anthropogenic radionuclides in the Ob River system. Our focus is on the water column because the transport and fate of particle-reactive radionuclides will be controlled by the transport of colloidal and suspended particles. Colloidal particles (1-200 nm), due to their large surface area, may be an especially effective mode of radionuclide transport in the low salinity region of the system. The aggregation of colloids at higher salinities will produce larger particles whose transport and deposition will differ from colloids. Moreover, ion exchange effects as colloidal and suspended particles are transported from lower to higher salinity may be important in controlling the transport of radionuclides such as ^{137}Cs. Our water column work has been done in conjunction with the field efforts organized in the Ob by Drs. Hugh Livingston and Fred Sayles of the Woods Hole Oceanographic Institution and complements their studies of bottom sediments (Sayles et al. 1997). The 1995 expedition followed a similar sampling trip in 1994 (reported in Final Report ONR Grant #N000149410967). We report here the 1995 data in tabular form, but discuss the 1994 and 1995 data sets together.

II. SAMPLE COLLECTION IN THE OB RIVER

A. Station locations

Water samples were collected in the Ob, Irtysh, and Tobal Rivers, in northwestern Siberia, from June 5 to June 22, 1995 (Table 1, Fig. 1). Sampling stations were located between the Arctic Circle and 58° North latitude, approximately from the town of Salekhard to just south of Tobolsk. The Russian Fisheries Protection vessel RS300#168, based in Salekhard, was used for the expedition. The water column sampling was successful, although the cruise was marred by the tragic death of Mr. Gera Panteleyev, a graduate student from the Woods Hole Oceanographic Institution who served as cruise coordinator and chief scientist.

Samples collected during this expedition include: barrels of filtered water for radionuclide analysis; filters and adsorber cartridges for analysis of particulate and dissolved radionuclides; particulate, dissolved, and whole water trace metal samples; filtered water for nutrient, DOC, and ^{129}I analyses; and suspended particulates for SPM, POC/PON, and chlorophyll analysis.

B. Radionuclides (Th, Am, Pu, Cs)

Large (approximately 57 l) filtered water samples were collected from 1-2 m depths at all stations. An attempt was made to maintain a sample depth of 2 m; however, the fast current caused the hose to ride up near the surface at some locations.

Filtered water was collected in 57 l barrels by pumping water through two 0.5 μm cartridge filters, designated Prefilter A and Prefilter B. After the barrel had been filled with filtered river water, pumping was halted. Two MnO_2 and two KFeCN cartridges were placed in series after the 0.5 μm prefilters, and pumping resumed, with the exit flow going to waste. The adsorber cartridges retain ^{137}Cs, Pu and Am.

C. Trace Metals

Whole water, dissolved and particulate trace metal samples were collected at nine stations. All water collected for trace metal analysis was hand-dipped from the bow of WHOI's inflatable catamaran, while the catamaran was motoring slowly forward through the water.

Trace metal vacuum filtrations were conducted at the end of each sampling day or the subsequent day, after equipment for other operations had been stowed. Due to the high suspended matter concentrations, 130 ml was about the maximum volume that could be passed through the precleaned 0.2 or 0.4 5m Nuclepore membranes. All trace metal clean equipment was stored in double plastic bags in an aluminum shipping box when not in use. The acid-cleaned polysulfone filtration assembly was rinsed with a water sample from each site prior to use, but was not acid-cleaned between sites. Several blanks were collected by filtration of Milli-Q water to evaluate the cleanliness of the work environment.

D. Nutrients, DOC, [129]I

Individual samples of filtered water were obtained for nutrient (phosphate, nitrate, nitrite, silicate), dissolved organic carbon (DOC), and [129]I analysis by pumping river water through a 0.2 μm Gelman capsule filter. About 5 l of river water were filtered through each Gelman filter. Nutrient samples were acidified with 0.1 ml of concentrated HCl. DOC samples were acidified with 0.5 ml of 50% H_3PO_4 and stored in the dark.

E. POC/PON

Surface water was collected from either the catamaran or by bucket from the vessel, and was filtered for particulate organic carbon and nitrogen (POC/PON). The volume filtered was always about 100 ml, and the filtrate volume was measured with a graduated cylinder after filtration. Precombusted 25 mm glass fiber filters (GF/F) were used on a plastic filter assembly. Two GF/F replicates were made, and both were placed in petri dishes and then wrapped in foil, so that either could be analyzed for POC/PON or chlorophyll.

F. Cross-flow filtration

At three stations, a cross-flow filtration system was used to obtain the truly dissolved (<1000 MW) size fraction for [129]I and DOC analysis. These samples were collected in two 500 ml plastic bottles.

G. Cruise participants

The Chief Scientist for the OB95 Expedition was Gera Panteleyev, a student at Woods Hole Oceanographic Institution (WHOI), although this role was taken over by Stephen Smith after Gera's untimely death. The PI's for WHOI's sediment radionuclide work are Dr. Fred Sayles and Dr. Hugh Livingston. Wendy Woods, a student at the University of Rhode Island (URI), conducted water column sampling for co-PI's Dr. S. Bradley Moran of URI and Dr. J. Kirk Cochran and Dr. Nicholas S. Fisher, of the State University of New York, Stony Brook. Other cruise participants included three Russian scientists: Dr. Olga Medkova (Russian Arctic and Antarctic Research Institute, St. Petersburg), and Alexei Moroshnikov and Nikolai Tarasov (Institute of Geology of Ore Deposits, Petrography, Mineralogy and Geochemistry, Moscow).

III. ANALYTICAL METHODS AND RESULTS

A. Large volume pumping

The prefilter cartridges from the large volume pumping retained ~10-50 g of suspended sediment (>0.5 μm) for analyses. These cartridges were ashed at 550°C for 24 hours and analyzed for ^{237}Np, ^{137}Cs and Pu isotopes, as follows:

^{237}Np and Pu isotopes were analyzed on ~1 g aliquots of the ashed sediment. These samples were supplied to Dr. T. Beasley of the Environmental Measurements Laboratory (New York, NY), who performed the radiochemical separations, followed by isotope dilution mass spectrometry. The latter was carried out by Dr. J. Kelley at Batelle Pacific Northwest Laboratories, (Richland, WA) according to procedures established at Batelle PNL. These data are reported in Tables 2 and 3.

Preliminary radiochemical analyses of Pu and ^{137}Cs on the MnO_2 and Cu(Fe)CN impregnated cartridges respectively, that followed the pre-filters in the large volume samples showed poor retention and no further analyses of these fractions were attempted.

^{137}Cs was measured by non-destructive gamma spectrometry of the cartridge ash using a low background intrinsic germainium detector. The detector was calibrated with NIST Standard Reference Material #1645 (River Sediment), measured in the same geometry as for the samples. The relatively high gamma activity of ^{137}Cs makes sample self-absorption corrections unnecessary. The standard was measured before and after each group of samples, but not less often than once per month. The ^{137}Cs activity are given in Table 4, as are $^{239,240}Pu$ activities calculated from the ^{239}Pu and ^{240}Pu atom data (Table 2). Uncertainties are 1σ counting errors.

B. Colloidal size-fractionated sampling

Aliquots of the fractions resulting from the cross-flow filtration of water samples were provided to Dr. L. Kilius of the Isotrace Laboratory, University of Toronto, for measurement of ^{129}I by accelerator mass spectrometry. The ^{129}I data are given in Table 5. Aliquots of these fractions also were analyzed at Stony Brook for DOC using a Shimadzu 5000 TOC analyzer.

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DOC values in the colloidal (1000 NMW - 0.2 μm) and truly dissolved (<1000 NMW) were high and variable, suggesting contamination by the cross-flow filtration unit. Only the <0.2 μm DOC values are reported here (Table 6). Uncertainty based on replicate measurements is estimated at ± 2%.

C. Ancillary samples

POC and PON were measured on glass fiber filter samples using a combustion method. The filters were dried at 60°C for 24 hours and fumed with concentrated HCl prior to CHN analysis. Quarter filter samples were combusted in a Carlo Erba EA 1108 Elemental Analyzer. Blank estimates were made using both clean filters and those subjected to HCl fuming. Precision is estimated to be ± 5%. The data are given in Table 6.

IV. DISCUSSION OF FIELD DATA

A. Pu isotopes, ^{137}Cs and ^{237}Np

Combining the data from the two years of sampling in the Ob River (1994, 1995) permits an assessment of trends in radionuclide distributions throughout the system. It is important to keep in mind however, that interannual variability may obscure some trends.

The specific activities (mBq/q) of ^{137}Cs and ^{239,240}Pu vary by about an order of magnitude through the system (Figs. 2,3). There is no clear trend in activities, although both Pu and Cs activities are notably greater in the samples taken in the Ob delta (94-11) and in the Taz (94-13). Specific activities can be affected by the nature of sediment in suspension and grain size (surface area) is an important factor. A more useful approach to determine potential sources of radionuclides is the activity ratio ^{239,240}Pu/^{137}Cs (Fig. 4). The sources of Pu and ^{137}Cs to the Ob system are several, including global fallout, release from the Mayak (Chelyabinsk) and Tomsk reprocessing facilities and fallout from the Semipalatinsk weapons tests. Releases from Mayak can make their way to the Ob via the Techa, Iset, Tobal and Irtysh Rivers. Tomsk is on the upper Ob and Semipalatinsk tests could have affected the drainage basins of the Irtysh and Ob (Fig. 1). The Pu and Cs released from these different sources likely have distinctive isotopic signatures. For example, the ^{239,240}Pu/^{137}Cs activity ratio of integrated global fallout in the northern hemisphere is ~0.027 (± .001) (corrected to 1994-95; Beck and Krey 1983). In constrast, the ^{239,240}Pu/^{137}Cs ratio of sediments in the Techa River is reported to be low (0.003 ± 0.002; Trapeznikor et al. 1993), as a consequence of releases from Mayak.

Suspended sediment samples collected south of the Arctic Circle in both 1994 and 1995 display the fallout ^{239,240}Pu/^{137}Cs activity ratio, within 2σ uncertainty. North of the Arctic Circle, in the delta region of the Ob and in the Taz River, greater values are observed. The latter, greater-than-fallout ratio in the Taz, is consistent with the results of Sayles et al. (1997) who noted slightly elevated ratios in a core taken in the Taz. Sayles et al. (1997) also point out the slight elevations in the ratio observed in lakes elsewhere, suggestive of some fractionation in the scavenging of Pu and Cs onto particle surfaces.

In contrast to the pattern of fallout or greater-than-fallout ratios in the Ob, the samples taken in the Irtysh and Tobal (with one exception, 95-7) all display values significantly below the global fallout value. These could be produced by a fraction of Mayak-labelled low Pu/Cs

sediment. We have no simple source explanation for the unusually high ratios at stations 95-7 and 94-8,10 except to note that low ^{137}Cs at the former and high ^{239,240}Pu activities at the latter stations seem responsible for these unusual ratios.

The ^{240}Pu/^{239,}Pu atom ratio is another indicator of radionuclide releases to the Ob (Fig. 5). Unlike the ^{239,240}Pu/^{137}Cs ratio, the ^{240}Pu/^{239,}Pu ratio should not be affected by differential chemical behavior, since isotopes of the same element are used. The global fallout value of this ratio is ~0.18 whereas Pu released from fuel processing has atom ratios of 0.04-0.06. The suspended sediments of the Ob system generally show ^{240}Pu/^{239,}Pu ratio less than the global fallout value of 0.18. The exceptions to the pattern are a closed pond at Salekhard (95-0) and the sample taken in the Taz River (94-13). Significantly, the values taken in the Ob River before its confluence with the Irtysh (95-4,5) are the lowest measured (0.126) suggesting that Pu from the Tomsk is making its way into the system. Values in the Irtysh are also depressed. The Tobal, which might be expected to show releases of Pu from Mayak, has ratios close to fallout (Table 7). If we use the ^{240}Pu/^{239}Pu atom ratio of fallout (0.18) and fuel reprocessing (0.04) as end numbers, we calculate that 39% of the Pu in suspended sediments of the Ob prior to its confluence with the Irtysh is from reprocessing sources. Elsewhere in the system, <20% of the Pu is added from these sources.

Both positive and negative departures from the global fallout value of ^{237}Np/^{239}Pu (0.45; Beasley et al. 1996) are evident in the Ob system (Fig. 6, Table 8). The ^{237}Np/^{239}Pu atom ratio of one sample taken in the Irtysh is markedly greater than fallout, but this is inconsistent with Np released from low yield weapons tests that might have affected the watershed of the Irtysh (Beasley, pers. comm.). Moreover, the ^{239}Pu atom concentration in that sample is the lowest observed in any of the suspended sediment samples collected in 1994 or 1995. The ^{237}Np, although among the lowest values, is not unusually low. Elsewhere in the Ob system, ^{237}Np/^{239}Pu ratios equal to or less than the fallout ratio are observed. It is possible that the low ratios are caused by lower uptake of Np relative to Pu onto particles.

B. Iodine-129

The pattern of ^{129}I in the Ob River system provides clear evidence of the release of this radionuclide from the Mayak reprocessing facilities (Fig. 7). "Dissolved" ^{129}I shows highest concentrations (~8 x 10^9 atoms/l) in the Tobal River before it joins the Irtysh. Our sampling of the colloidal and truly dissolved (<1 kD) fractions shows that a high percentage (>70%) of the ^{129}I that passes a 0.2 μm filter is truly dissolved in most of the Ob system. In contrast to the Tobal results, dissolved ^{129}I concentrations in the Irtysh above its confluence with the Tobal and in the Ob above its confluence with the Irtysh are low, ~2 x 10^9 and ~1 x 10^9 atoms/l respectively. As these rivers merge, first with the Tobal and then with the Irtysh, the Mayak ^{129}I signal is progressively diluted. Comparison of the concentrations with flow volumes will be undertaken to determine if the ^{129}I behaves conservatively during mixing. North of the confluence of the Ob and Irtysh, there is little change in ^{129}I concentration (values range from 2.2-2.8 x 10^9 atoms/l) until the broad delta portion of the system is reached (station 94-11,14,16). The lowest concentrations are measured in the Taz River (0.7 x 10^9 atoms/l). It is possible that some of the decrease in ^{129}I in the lower Ob is due to removal of I from solution. Concentrations of dissolved organic carbon also decrease in this region (Fig. 8).

V. LABORATORY EXPERIMENTS OF RADIONUCLIDE PARTICLE REACTIVITY

A series of laboratory experiments was conducted to assess the particle-reactivity of ^{241}Am, ^{137}Cs, ^{57}Co (as an analogue of ^{60}Co), and ^{131}I (as an analogue for ^{129}I) in the Ob. These experiments were designed to help understand how these radionuclides would behave if they were transported in the Ob to the saline portion of the estuary. Specifically, would these radionuclides associate with particulate matter or form particles once they reached saline waters? Moreover, to what extent is their behavior influenced by the high concentrations of dissolved organic matter in the Ob water?

Experiments were conducted as follows: Sterile-filtered (0.2 μm) Ob River water (at 0 ppt salinity) received pM to nM levels of the radioisotope elements and was mixed with different amounts of sterile-filtered seawater (collected 8 km off Southampton, NY) to produce final salinities of 0, 1, 3, or 5 ppt salinity. This water was incubated at 2 C for several days, and periodically samples were taken to determine the particle-association of the radionuclides, using either 0.2 or 1 μm Nuclepore polycarbonate membranes following established filtration techniques (Fisher et al. 1983). The fractionation of each radionuclide between dissolved and particulate phases was determined at each sample time; the results shown below reflect their fractionations at time of apparent equilibrium between dissolved and particulate phases. This work was repeated with filtered Ob water that was UV-irradiated to phtooxidize the dissolved organic matter. The overall experiment was repeated several times, each time with triplicate samples.

Representative results are shown in Figs. 9 and 10. Overall, the particle-reactivity of the radionuclides was in the order Am>>>Co=I≥Cs. The association of the radioisotopes with particles reached equilibrium within 1-2 days; equilibrium values for all radioisotopes are given in Table 8. For Am and, to a lesser extent Co, the particle association (> 0.2 μm) increased with salinity (from 0 to 5 ppt) (Fig. 9). Thus, for Am, the fraction associated with particulate matter >0.2 μm increased from 1.1% at 0 ppt salinity to 37.6% at 5 ppt. The fraction associated with particles >1 μm increased from 0.3% at 0 ppt salinity to 4.2% at 5 ppt. Thus most of the particulate matter with which the Am was associated was larger than 0.2 μm but smaller than 1 μm. In contrast, there were no significant differences between 0.2 and 1 μm for Co, except at 5 ppt. However it is noteworthy that even when it was most particle reactive (at 5 ppt), < 0.3% of the Co was associated with particulate matter (Fig. 9). There was an opposite trend for Cs and I with respect to particle reactivity as a function of salinity. Again, these elements are extremely unreactive for particulate matter (values typically < 0.3% on particles), but Cs and I were most particle reactive at 0 ppt salinity and reactivity decreased with increasing salinity (Fig. 9).

When the dissolved organic matter in the Ob water was photooxidized by high intensity UV irradiation, the particle reactivity at 0 ppt salinity of Am increased from 1.1% (> 0.2 μm) in unirradiated water to 16.0% in irradiated water (or from 0.3% to 5.8%, > 1 μm) (Table 8, Fig. 10). UV irradiation also increased the particle reactivity of Co (> 0.2 μm), but had no effect on the particle reactivity of Cs or I (Fig. 10).

Overall, it can be concluded that of the radionuclides examined, only Am is likely to display significant "salting out" effects in the saline portion of the Ob estuary. The high numbers of

competing ions and stable Cs and I in saline water actually decreased the particle association of these elements, which are not very particle reactive in aquatic systems. Thus, these other radionuclides would be expected to be transported readily throughout the estuary, whereas Am may be deposited in sediment near where the saline water meets the inflowing river water. Moreover, it is apparent that the high amounts of dissolved organic matter in the Ob are effective in helping to transport Am to the Kara Sea. The particle association of Am at increasing salinity may well be due to the coagulation of dissolved organic matter (particularly in the size range of 0.2 - 1 μm), some of which is complexed with Am.

VI. STATISTICAL INFORMATION

A. Publications and Abstracts
Fisher, N.S., S.W. Fowler, F. Boisson, J.L. Carroll, K. Rissanen, B. Salbu, T.G. Sazykina and K.L. Sjoebtom. 1997. Bioconcentration factors and sediment partition coefficients for radionuclides in contaminated Arctic Seas. Env. Sci. Tech., submitted.

Livingston, H.D., F. Sayles, J.K. Cochran, S.B. Moran and T. Beasley. 1996. Transport of Pu isotopes from various sources in the Ob River system during the last 50 years. EOS, Trans. Am. Geophys. Un., 77(46), p. F376.

Moran, S.B., J.K. Cochran, N.S. Fisher and L.R. Kilius. 1995. [129]I in the Ob River. In: Proceedings of the Second International Conference on Environmental Radioactivity in the Arctic. Oslo, Norway, August 21-25, pp. 75-78.

Moran, S.B. and W.L. Woods. 1996. Cd, Cr, Cu, Ni and Pb in the water column and sediments of the Ob-Irtysh Rivers, Russia. Marine Pollution Bulletin (in press).

B. Presentations
Cochran, J.K., ANWAP Workshop on Risk Assessment, "Transport and fate of anthropogenic radionuclides in the Ob River system", Sequim, WA, October, 1995.

Fisher, N., ANWAP Workshop on Risk Assessment, "Bioconcentration factors and sediment Kds' of long-lived radionuclides in the Ob River and Kara Sea", Sequim, WA, October 1995.

Fisher, N., ANWAP Workshop, "Transport and fate of anthropogenic radionuclides in the Ob River system", Snowbird, UT, May 1996.

C. Graduate Students supported
Mr. Gabrial Kra, SUNY-Stony Brook
Ms. Wendy Woods, University of Rhode Island

D. <u>Undergraduate Students supported</u>
Michael Boyer, Harvard Univ.
Christopher Hoimes, SUNY-Stony Brook

E. <u>Post-doctoral Fellows supported</u>
David Hutchins
Wen Wang

VII. REFERENCES

Beasley, T.M., J.M. Kelley, T.C. Maiti and L.A. Bond (1996) ^{237}Np/^{239}Pu ratios in integrated fallout: a reassessment of the production of ^{237}Np. Chem. Geol. Isot. Geosci., in press.

Beck, H.L. and P.W. Krey (1983) Radiation exposure in Utah from Nevada nuclear tests. <u>Science</u> 220, 18-24.

Fisher, N.S., P. Bjerregaard and S.W. Fowler (1983) Interactions of marine plankton with transuranic elements. I. Biokinetics of neptunium, plutonium, americum, and californium in phytoplankton. <u>Limnol. Oceanogr.</u> 28, 432-447.

Sayles, F.L., H.D. Livingston and G.P. Panteleyev (1997) The history and source of particulate ^{137}Cs and ^{239,240}Pu deposition in sediments of the Ob River delta, Siberia. <u>Sci. Tot. Euv.</u>, in press.

Trapeznikov, A.V., A. Aarkrog, N.V. Kulikov, S.P. Nielsen, V.N. Pozolotina, G. Polikarpov, V.N. Trapezhikova, M. Ya Chebotina, and P. Yushkov (1993) Radioactive contamination of the Ob River system from the nuclear enterprise "Mayak" in the Urals. Proc. Intl. Conf. Environmental Radioactivity in the Arctic and Antarctic, 23-27 Aug. 1993, Kirkines, Norway.

List of Figures

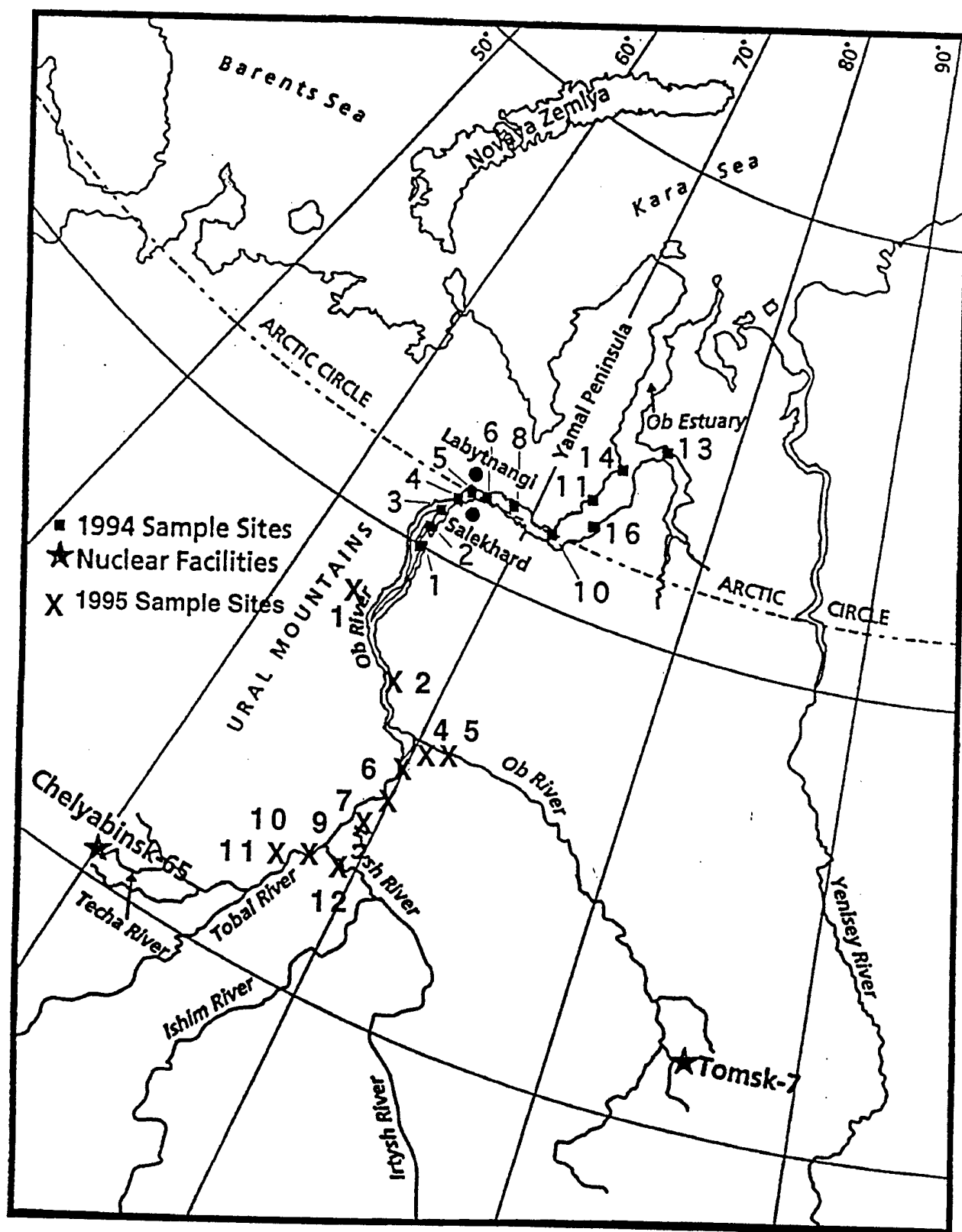Fig. 1. Map showing stations sampled in the Ob River system. Stations marked with a filled square were sampled in 1994. Samples indicated with an "x" were sampled in 1995.
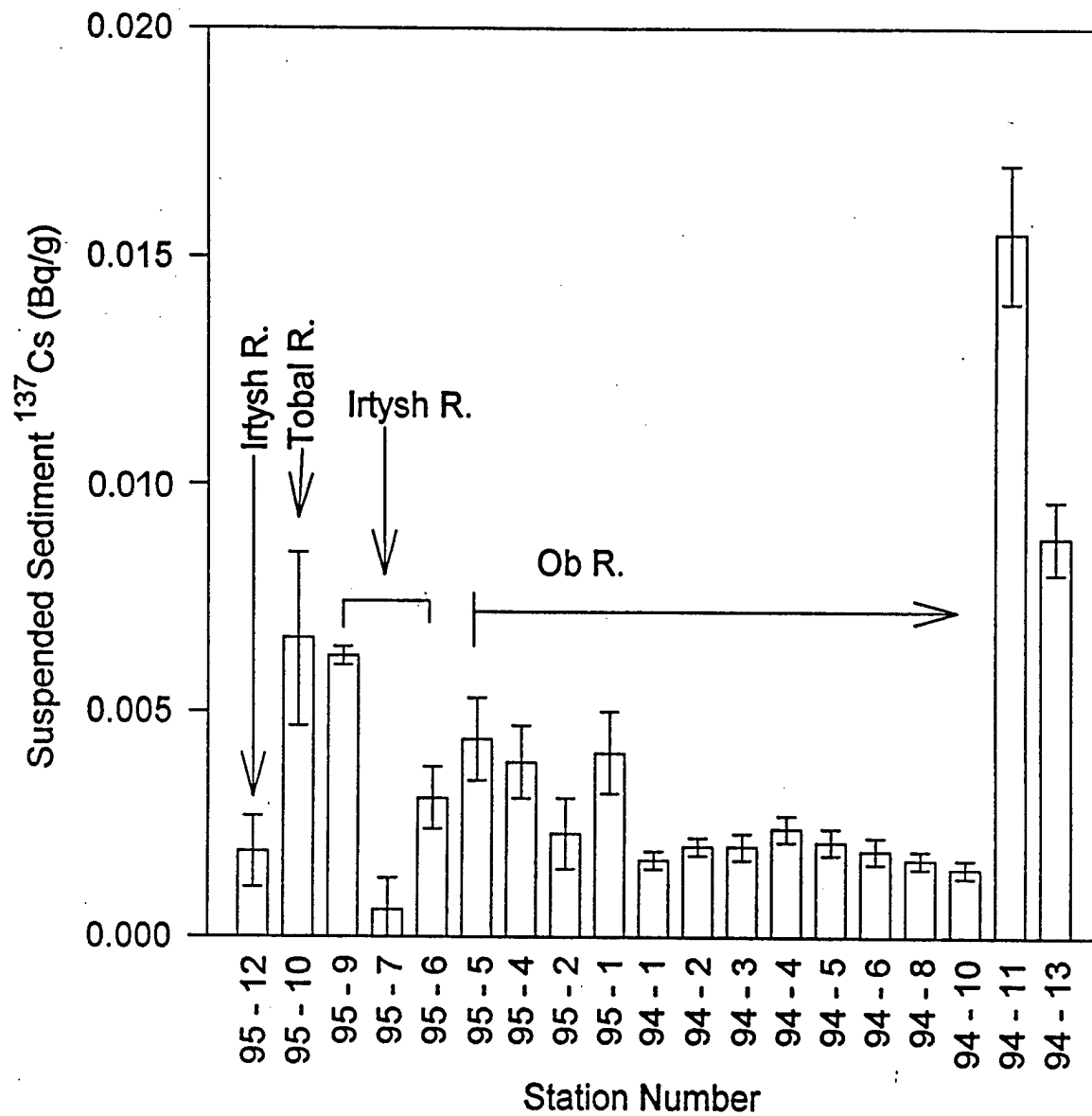
Fig. 2. Specific activities (Bq/g) of ^{137}Cs in suspended sediment (>0.5 μm) of the Ob River system.
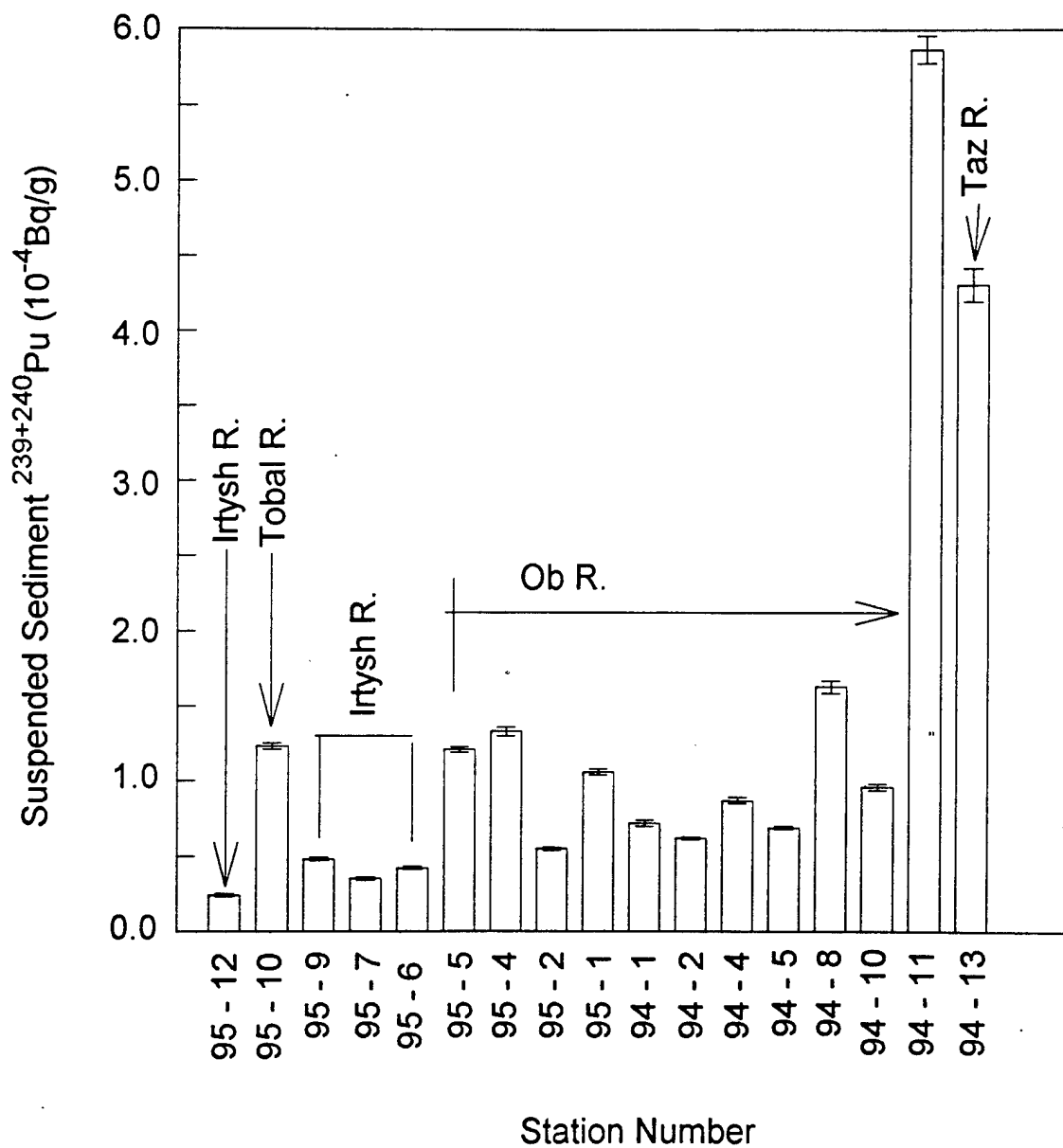
Fig. 3.   Specific activities (10^{-4} Bq/g) of ^{239,240}Pu in suspended sediment (>0.5 μm) of the Ob River system.
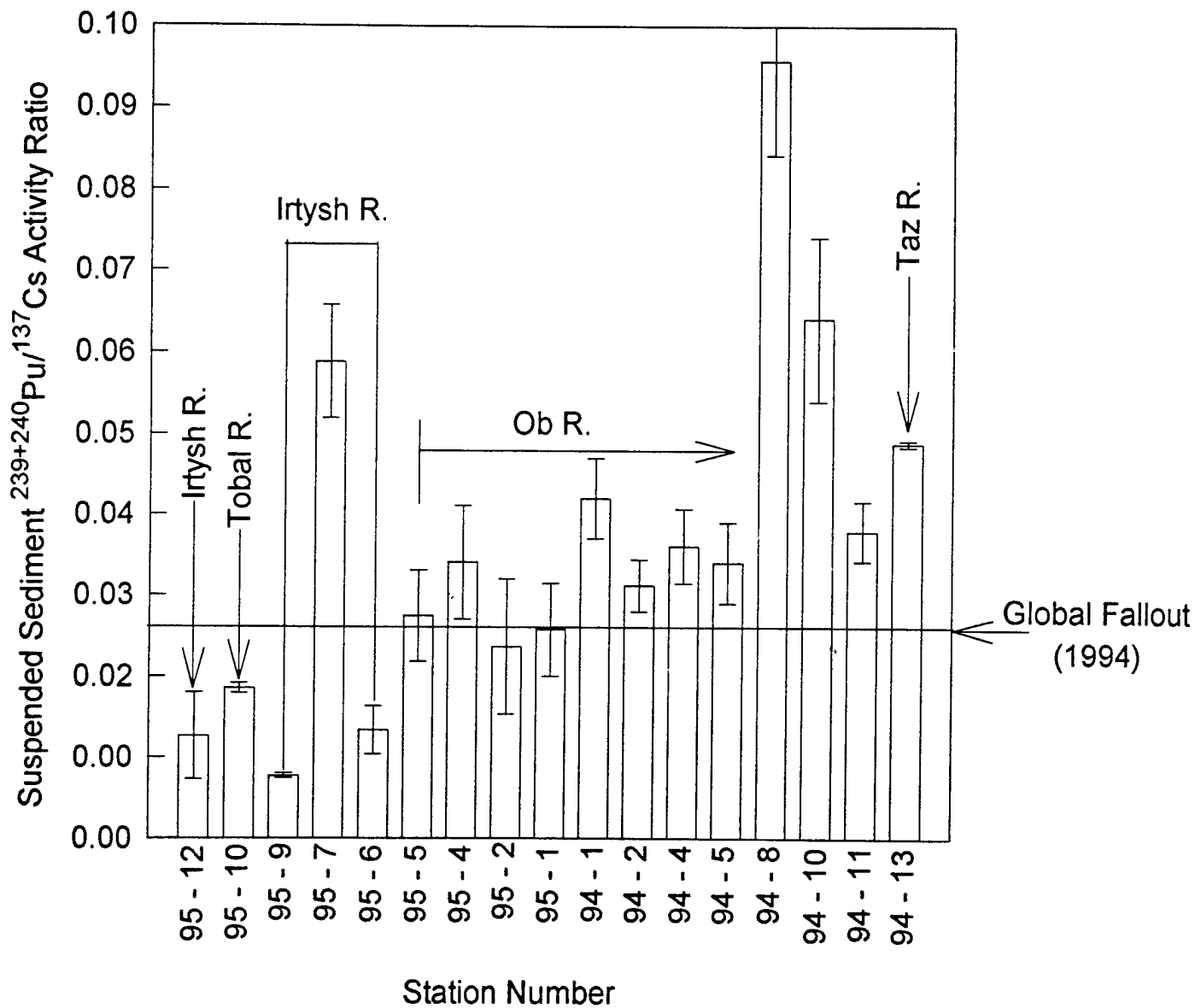
Fig. 4. ^{239,240}Pu/^{137}Cs activity ratio of suspended sediment in the Ob River system.
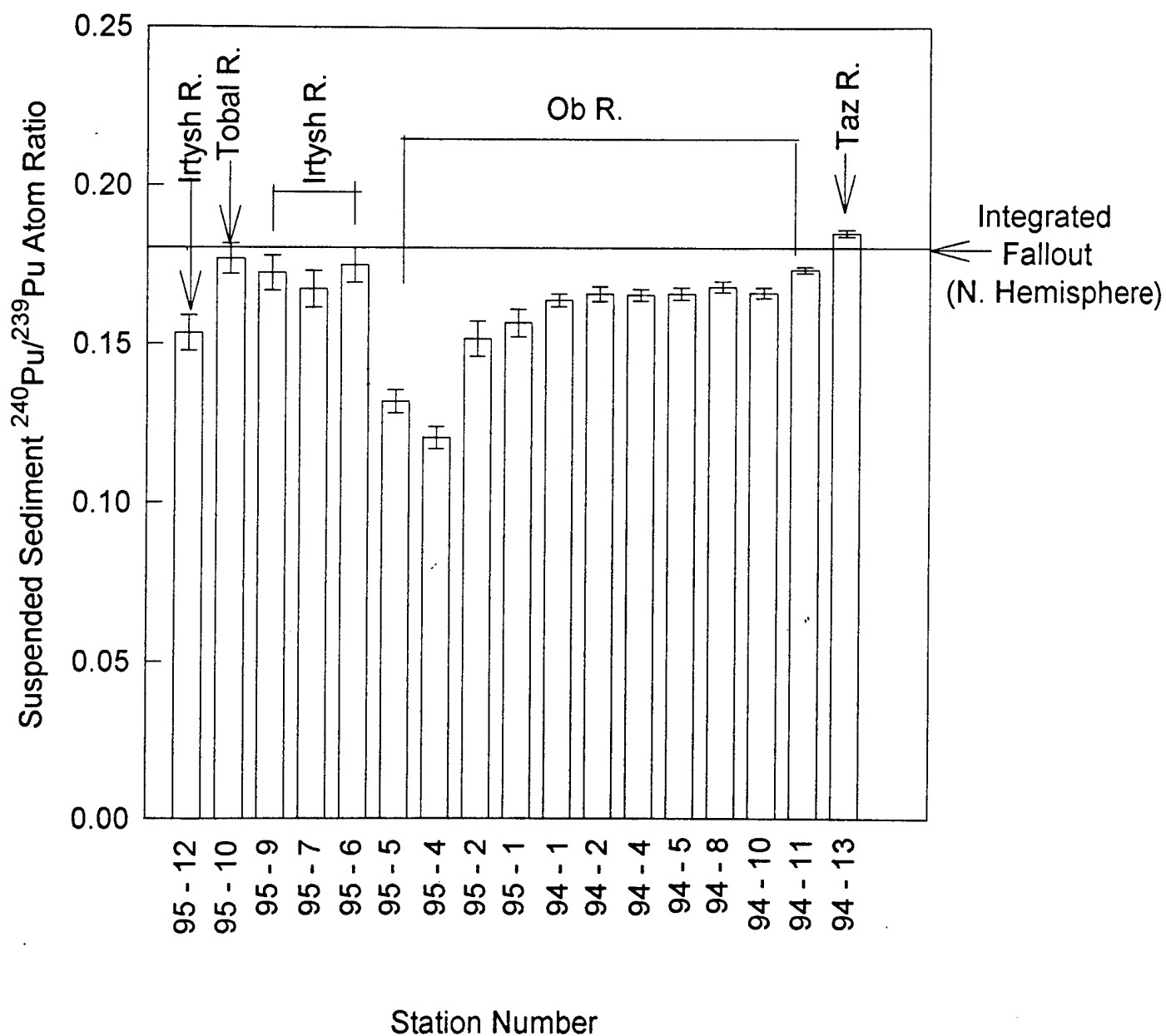
Fig. 5. ^{240}Pu/^{239}Pu atom ratio in suspended sediment (>0.5 μm) in the Ob River system.
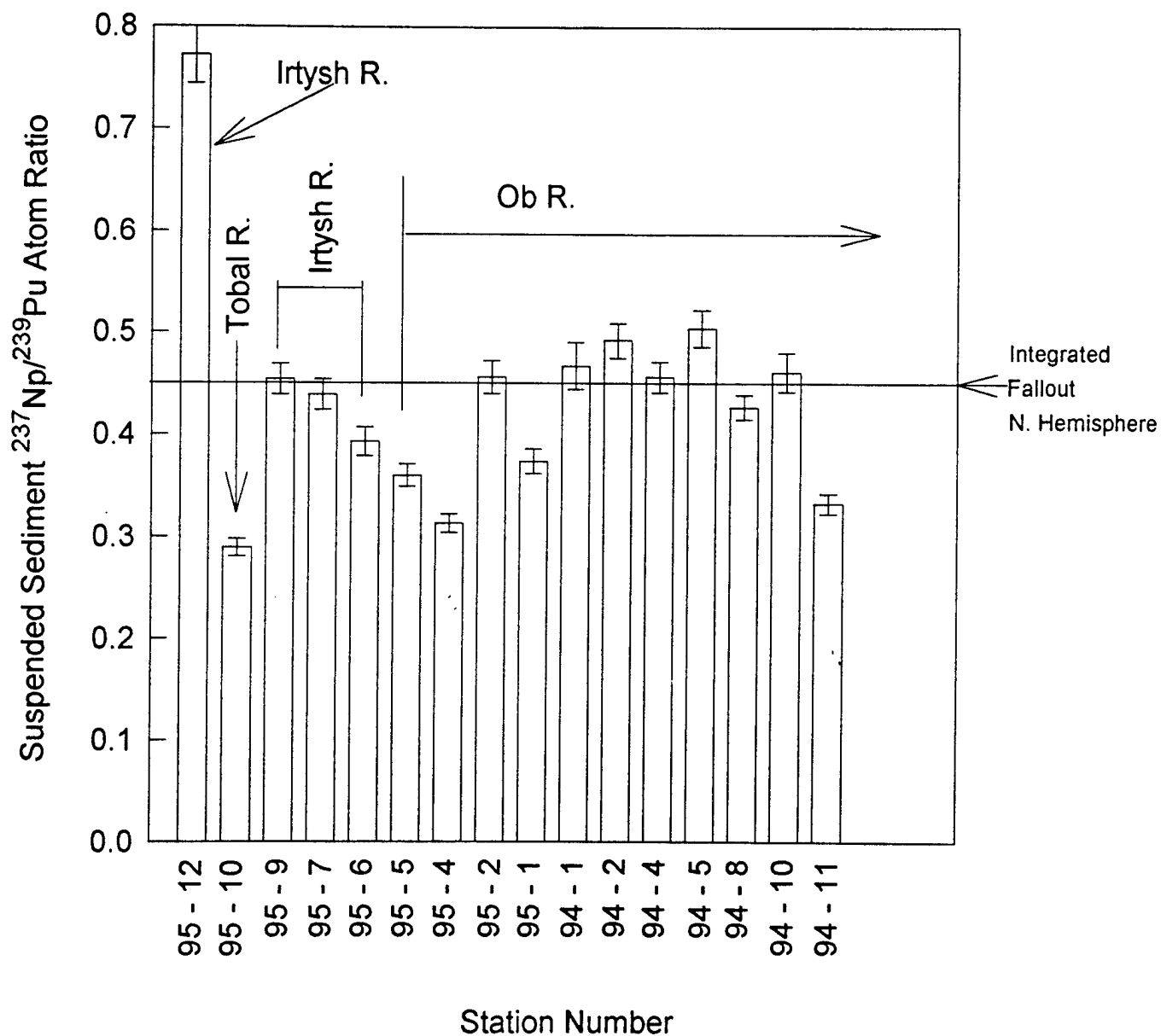
Fig. 6. ^{237}Np/^{239}Pu atom ratio in suspended sediment (>0.5 μm) in the Ob River system.

Ob River I-129
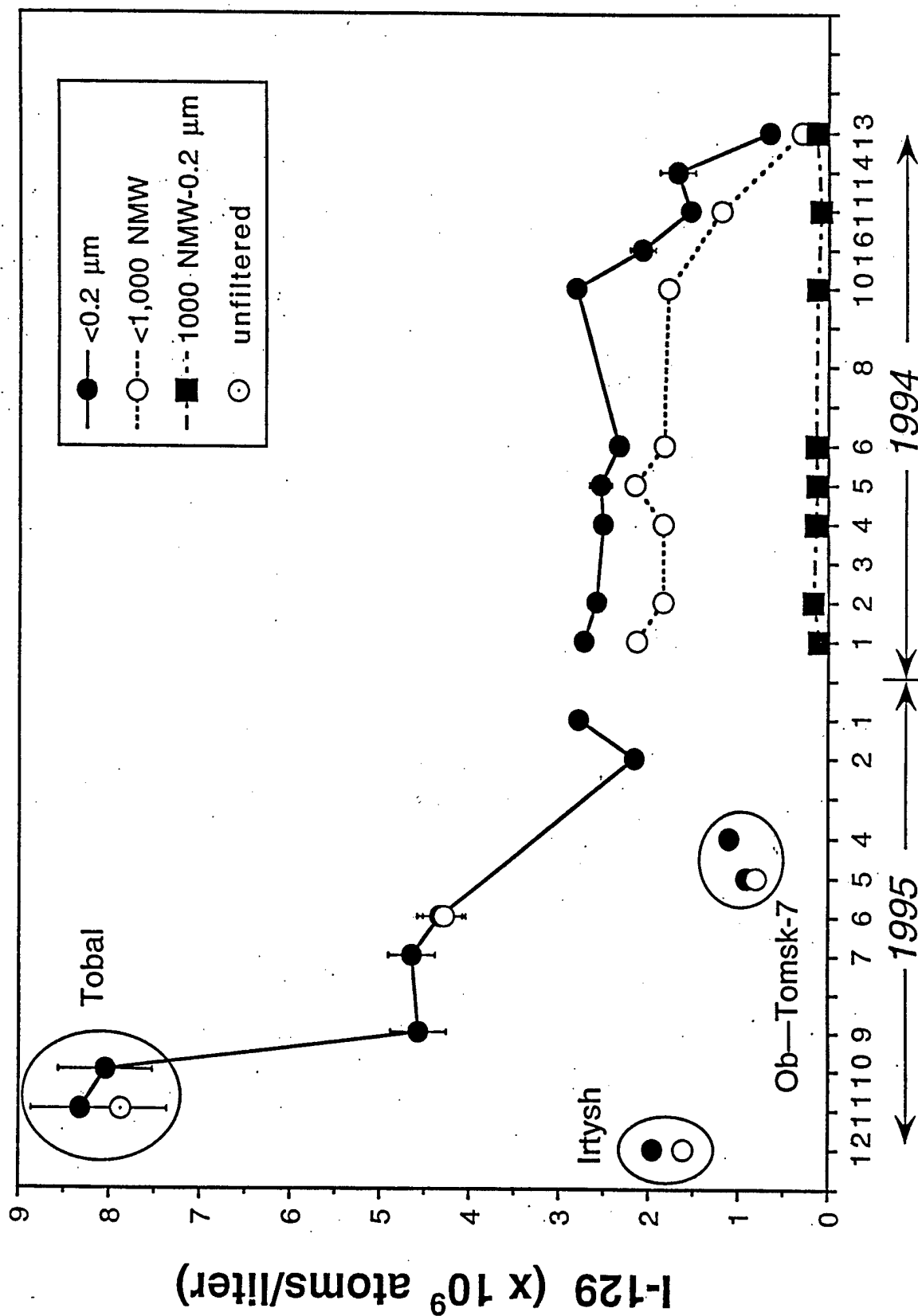
Fig. 7. [129]I in size-fractionated samples of Ob River water. Samples were filtered through 0.2 μm filters to collect "dissolved" I then through cross-flow filtration to separate colloidal [129]I (1000 NMW - 0.2 μm) from truly dissolved (<1000 NMW).
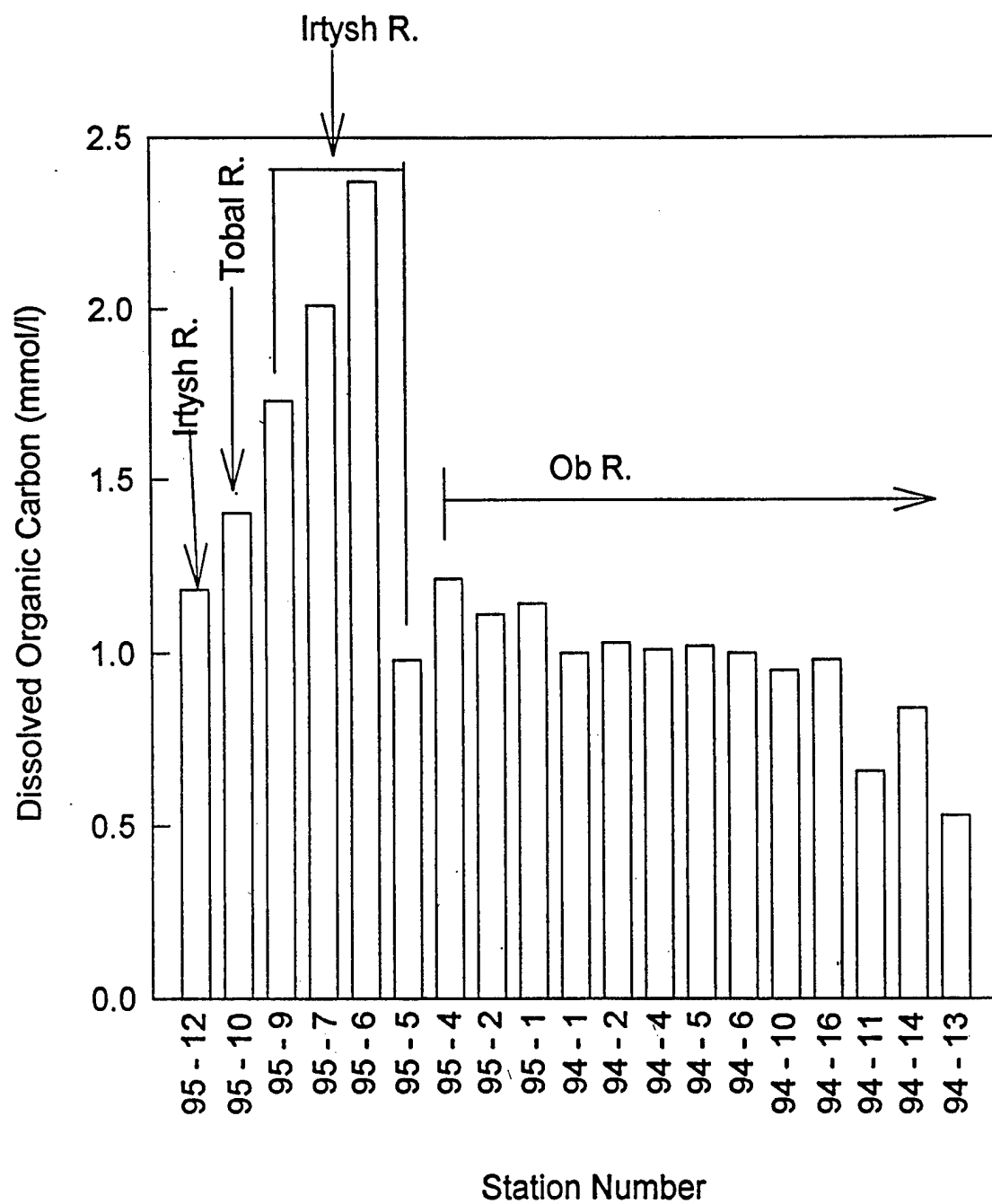
Fig. 8. Dissolved organic carbon (<0.2 μm) in the Ob River system.
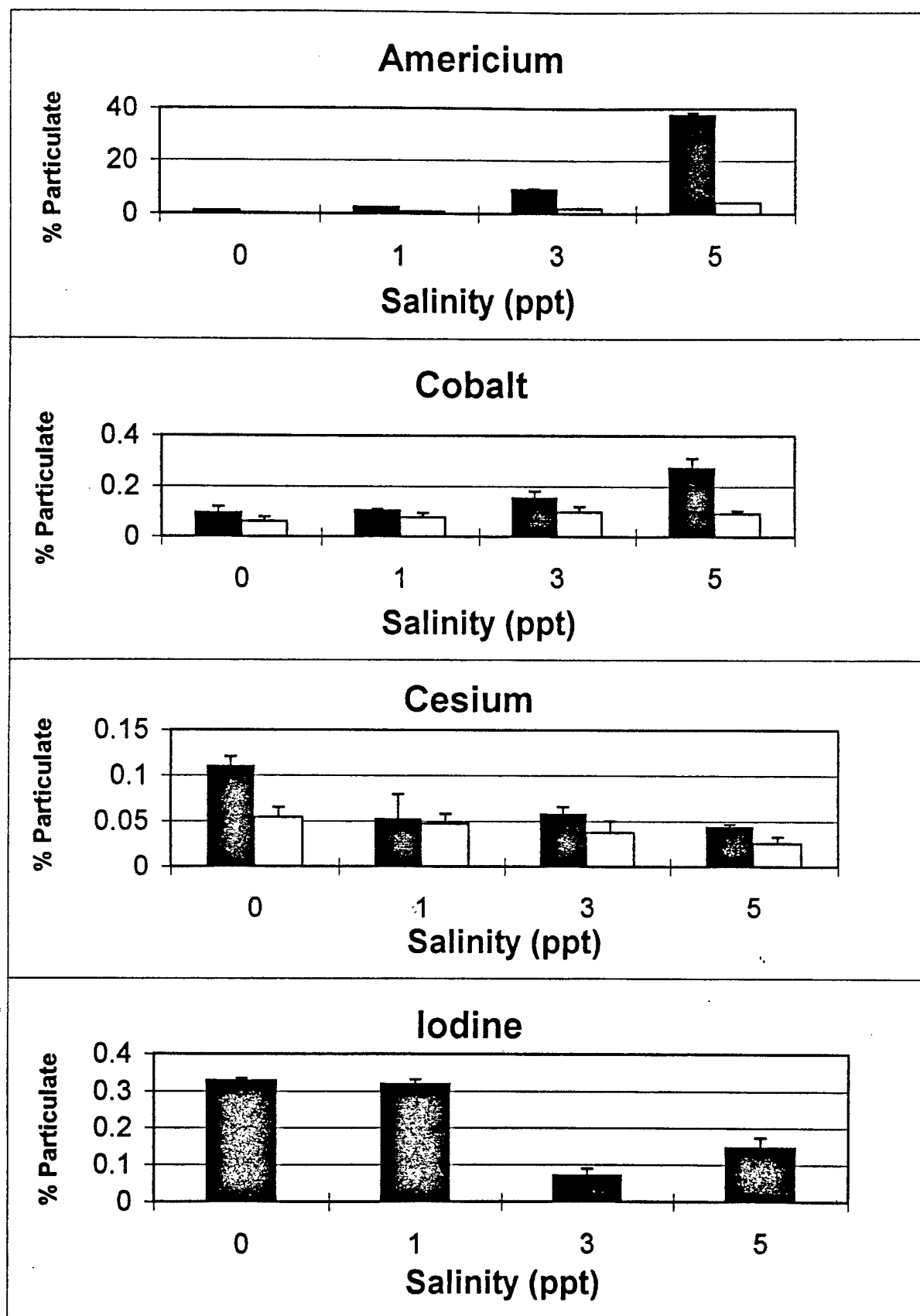
Fig. 9. Effects of salinity on the particle-reactivity of Am, Co, Cs, and I in Ob river water. Solid bars: filtered through 0.2 μm membranes; open bars: filtered through 1 μm membranes. Data are means of three replicates ± standard deviation. For I, only 0.2 μm membranes were used.
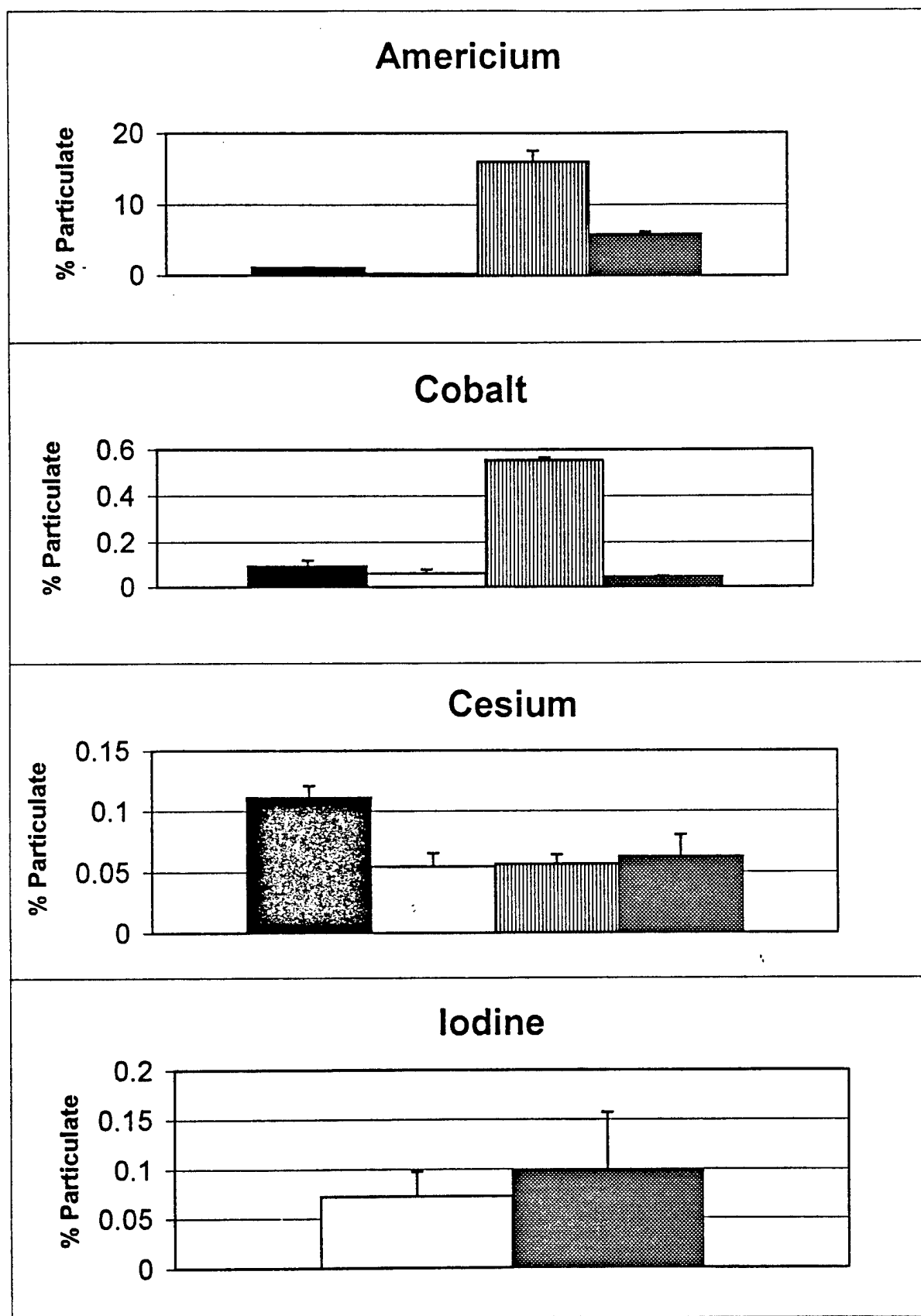
Fig. 10. Effects of UV irradiation on the particle-reactivity of Am, Co, Cs, and I in Ob River water at 0 ppt salinity. (■): unirradiated Ob water, filtered through 0.2 μm; (□): unirradiated Ob water, filtered through 1μm; (▥): UV-irradiated Ob water, filtered through 0.2 μm; (▨): UV-irradiated Ob water, filtered through 1 μm. Data are means of three replicates ± 1 standard deviation. For I, only 1μm membranes were used.

Table 1: Ob River station locations and ancillary data.

Station	Date (mm/dd/yy)	Location	
95-0	06/05/95	Arctic Circle, Salekhard	
95-1	06/07/95	63°35.303'N	65°9.592'E
95-2	06/11/95	62°26.702'N	66°1.862'E
95-4	06/13/95	61°11.268'N	68°55.487'E
95-5	06/13/95	61°11.493'N	69°4.433'E
95-6	06/14/95	60°56.635'N	69°12.759'E
95-7	06/16/95	59°34.174'N	69°19.033'E
95-9	06/18/95	58°30.619'N	68°27.509'E
95-10	06/19/95	58°5.740'N	68°9.210'E
95-11	06/20/98	(see map, Fig. 1 for approximate location)	
95-12	06/21/95	57°59.628'N	68°48.289'E

Table 2: Radionuclide data for suspended sediments of the Ob River (1995).

Station	^{237}Np (x10^7)	^{239}Pu (x10^7)	^{240}Pu (x10^6)	^{241}Pu* (x10^4)	^{242}Pu (x10^5)
		------------------------------------atoms/g sed------------------------------------				
95-0	5.74 ± 0.17	25.87 ± 0.46	47.09 ± 0.88	72.5 ± 1.9	10.34 ± 0.26
95-1	2.757 ± 0.089	7.37 ± 0.13	11.54 ± 0.23	18.6 ± 1.1	2.53 ± 0.15
95-2	1.752 ± 0.059	3.844 ± 0.084	5.83 ± 0.17	7.99 ± 0.9	0.99 ± 1.2
95-4	3.166 ± 0.092	10.11 ± 0.020	12.17 ± 0.26	16.3 ± 1.5	2.00 ± 0.15
95-5	3.21 ± 0.10	8.93 ± 0.16	11.77 ± 0.25	16.8 ± 1.0	2.03 ± 0.13
95-6	1.095 ± 0.038	2.784 ± 0.055	4.86 ± 0.12	6.41 ± 0.98	1.03 ± 0.10
95-7	1.053 ± 0.035	2.400 ± 0.049	4.01 ± 0.11	6.24 ± 0.67	0.81 ± 0.09
95-9	1.449 ± 0.048	3.194 ± 0.063	5.50 ± 0.14	6.48 ± 0.67	1.016 ± 0.089
95-10	2.359 ± 0.073	8.16 ± 0.15	14.42 ± 0.29	20.94 ± 1.2	3.10 ± 0.13
95-12	1.307 ± 0.047	1.694 ± 0.035	2.597 ± 0.077	3.05 ± 0.94	0.48 ± 0.09

*Decay corrected to 1 January 1995.
Uncertainties are 1σ errors.

Table 3: Radionuclide atom ratios for suspended sediments of the Ob River (1995).

Station	$^{237}Np/^{239}Pu$	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu*$ (x10^{-2})	$^{242}Pu/^{239}Pu$ (x10^{-2})
95-0	0.2217 ± 0.0065	0.18201 ± 0.00097	0.2803 ± 0.0056	0.3995 ± 0.0068
95-1	0.374 ± 0.012	0.1567 ± 0.0013	0.253 ± 0.014	0.343 ± 0.020
95-2	0.456 ± 0.016	0.1517 ± 0.0028	0.208 ± 0.023	0.257 ± 0.030
95-4	0.3130 ± 0.0092	0.1204 ± 0.0011	0.162 ± 0.015	0.198 ± 0.014
95-5	0.360 ± 0.011	0.1318 ± 0.0015	0.188 ± 0.011	0.226 ± 0.014
95-6	0.393 ± 0.014	0.1748 ± 0.0027	0.230 ± 0.035	0.371 ± 0.034
95-7	0.439 ± 0.015	0.1672 ± 0.0032	0.260 ± 0.027	0.335 ± 0.038
95-9	0.454 ± 0.015	0.1722 ± 0.0026	0.203 ± 0.021	0.318 ± 0.027
95-10	0.2890 ± 0.0087	0.1767 ± 0.0015	0.250 ± 0.014	0.380 ± 0.014
95-12	0.772 ± 0.028	0.1533 ± 0.0032	0.180 ± 0.055	0.283 ± 0.051

*Decay corrected to 1 January 1995.
Uncertainties are 1σ errors.

Table 4: Radionuclide activities and activity ratios for suspended sediments of the Ob River (1995).

Station	^{239,240}Pu* (mBq/g)	^{137}Cs (mBq/g)	(^{239,240}Pu/^{137}Cs)
95-0 (Salekhard)	0.39 ± 0.007	-	-
95-1	0.106 ± 0.002	4.10 ± 0.90	0.026 ± 0.006
95-2	0.055 ± 0.001	2.30 ± 0.80	0.024 ± 0.008
95-4	0.133 ± 0.003	3.90 ± 0.80	0.034 ± 0.007
95-5	0.121 ± 0.002	4.40 ± 0.90	0.028 ± 0.006
95-6	0.042 ± 0.001	3.10 ± 0.70	0.013 ± 0.003
95-7	0.035 ± 0.001	0.60 ± 0.70	0.059 ± 0.069
95-9	0.048 ± 0.001	0.62 ± 0.20	0.008 ± 0.001
95-10	0.123 ± 0.002	0.66 ± 0.80	0.019 ± 0.001
95-12	0.024 ± 0.001	1.90 ± 0.80	0.013 ± 0.005

*Atom concentrations for 239Pu and 240Pu (Table 2) converted to activities.
Uncertainties are 1σ errors.

Table 5: Water column [129]I results from the 1995 Ob River expedition.

| Station | Water column (10^9 atoms/l) | | |
	<0.2 μ ("Dissolved")	<1kD (Truly dissolved)	1kD - 0.2 μm (Colloidal)
95-1	2.78 ± 0.09	nm	nm
95-2	2.16 ± 0.008	nm	nm
95-4	1.10 ± 0.05	nm	nm
95-5	0.91 ± 0.04	0.80 ± 0.05	nm
95-6	4.33 ± 0.25	4.28 ± 0.24	nm
95-7	4.64 ± 0.26	nm	nm
95-9	4.57 ± 0.31	nm	nm
95-10	8.04 ± 0.52	nm	nm
95-11	8.32 ± 0.54	nm	nm
95-12	1.95 ± 0.07	1.61 ± 0.08	nm

nm = not measured

Table 6: Ob River DOC, POC and PON concentrations (1995).

Station	POC (μmol/l)	PON (μmol/l)	DOC (mmol/l)
95-1	-	-	1.14
95-2	269	5.82	1.11
95-4	105	2.67	1.21
95-5	83.9	1.20	0.98
95-6	134	2.55	2.37
95-7	77.3	2.00	2.01
95-9	99.9	3.49	1.73
95-10	139.2	6.25	1.40
95-12	82.9	2.84	1.18

Table 7: Isotopic ratios in the Ob River System.

Location	^{239,240}Pu/^{137}Cs (Bq/Bq)	^{240}Pu/^{239}Pu (atom/atom)	^{237}Np/^{239}Pu (atom/atom)	Potential Source
Tobal (n=1)	0.019	0.177±0.002	0.289	Mayak
Irtysh (n=1)	0.013±005	0.153±0.003	0.772±0.028	Semipalatinsk
Irtysh (north of confluence with Tobal; n=3)	0.027±0.028	0.171±0.004	0.429±0.032	-
Ob (south of confluence with Irtysh; n=2)	0.031	0.126	0.337	Tomsk
Ob (confluence with Irtysh to Salekhard; n=6)	0.032±007	0.162±006	0.458±0.046	-
Ob (north of Salekhard; n=3)	0.066±029	0.169±0.004	0.407±0.067	-
Taz (n=1)	0.049	0.185	0.232	-
Integrated northern hemisphere fallout	0.027	0.18	0.45	-

*Potential nonglobal source nearest to sampling sites.

**n = number of samples; uncertainty, where given, is 1σ of mean

Table 8. Fraction (%) of Am, Co, Cs, and I associated with particles >0.2 μm or 1 μm at time of apparent equilibrium (typically 48 h) in filtered, unirradiated Ob water mixed with varying amounts of filtered seawater to yield final salinities of 0, 1, 3, and 5 ppt. Also shown are fractionations of these elements in Ob water that was UV-irradiated and filtered before radioisotopes were added. All values are means of three replicates $\pm$ 1 standard deviation. nd: not determined.

Salinity (ppt)		Filter	Am	Co	Cs	I
0		0.2 μm	1.08 $\pm$ .11	0.09 $\pm$ .03	0.11 $\pm$ .01	0.33 $\pm$ .01
		1 μm	0.26 $\pm$ .08	0.06 $\pm$ .02	0.05 $\pm$ .01	0.07 $\pm$ .03
	UV	0.2 μm	16.0 $\pm$ 1.57	0.56 $\pm$ .01	0.56 $\pm$ .01	nd
	UV	1 μm	5.81 $\pm$ .34	0.04 $\pm$ .00	0.06 $\pm$ .02	0.10 $\pm$ .06
1		0.2 μm	2.38 $\pm$ .01	0.10 $\pm$ .02	0.05 $\pm$ .03	0.32 $\pm$ .01
		1 μm	0.66 $\pm$ .07	0.08 $\pm$ .02	0.05 $\pm$ .01	nd
3		0.2 μm	9.04 $\pm$ .42	0.15 $\pm$ .03	0.06 $\pm$ .01	0.07 $\pm$ .02
		1 μm	1.86 $\pm$ .20	0.10 $\pm$ .02	0.04 $\pm$ .01	nd
5		0.2 μm	37.6 $\pm$ .99	0.27 $\pm$ .04	0.04 $\pm$ .00	0.15 $\pm$ .03
		1 μm	4.22 $\pm$ .11	0.09 $\pm$ .01	0.03 $\pm$ .01	nd